Nonlinear Optics and Quantum Optics with X-Rays

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1 Nonlinear Optics

With the advent of 4th-generation x-ray sources, especially x-ray free-electron lasers (XFELs), x-ray optics will enter a qualitatively new regime. The most striking differences to state-of-the-art 3rd-generation sources are the transverse coherence and the photon degeneracy (photons per mode). Whereas a 3rd-generation source emits hard x-rays into hundreds of transverse modes and at a degeneracy of about 0.1, an XFEL will deliver 10^9 photons into a single transverse mode. This marks an important transition to nonlinear optics, where multiple photons are interacting coherently with each other through a suitable nonlinear medium: For a photon in a given mode to interact coherently with one in an another, the first requirement is to find one there. One can also consider incoherent nonlinear effects, such as incoherent two-photon absorption, but these are also limited by probability of finding a photon that can interact with a state generated by the previous absorption of a photon (see below). Therefore, nonlinear-optical effects are hard (but not impossible, see below) to find at a 3rd-generation source, but should be much more prominent at an XFEL. It still takes a suitable nonlinear medium to couple the interacting modes, and considerable theoretical and experimental effort will be required understand these. With that understanding, entirely new insights about matter can be obtained from nonlinear x-ray optics. X-rays and visible light interact differently with matter, and that makes nonlinear optics qualitatively different. With near-visible light, the optical response of a material is governed by the electrons in chemical bonds. Because x-ray-photon energies vastly exceed those of chemical bonds, the electrons in them can be considered free. Chemical bonds become important only at x-ray photon energies close to an absorption edge, i.e., a transition from an inner-shell electron to the valence structure. In some cases, nuclear resonances become important.

2 X-Ray Quantum Optics

X-ray quantum optics is a largely unexplored field - exceptions being the effects of parametric down conversion and nuclear γ-ray superradiance. To some extent, ideas and methods can be adapted from visible-light quantum optics, and be put to use for improved instrumentation and experiemntal techniques, such as x-ray spectroscopy (see below). This is becoming particularly relevant witht he advent of XFELs. However, there are a few crucial differences between visible and x-ray quantum optics, and these can be used to gain new insights:

- chemical bonds, which are responsible for visible-light interaction with matter are rather irrelevant; most interactions can be described in terms of almost-free and strongly bound (inner-shell) electrons, as well as nuclear γ-ray resonance
- photon detection efficiencies of much better than 99% are possible, which is important to close "loopholes" in tests of quantum physics
- as photon energies approach 1022 keV, virtual-pair interactions play an increasing role, i.e., the vacuum becomes different from that for visible-light photons. The typical photon energies of 10 to 30 keV delivered by 3rd and 4th-genration x-ray sources are a good step in this direction.

3 Estimates of the Strengths of Nonlinear-Optical Effects

We will first look at incoherent two-photon excitations with x-rays. This does not include processes where an intermediate relaxation process occurs. Although the participating photons need not be correlated with each other, the photon degeneracy is relevant because absorption of a second photon by the excited intermediate state is in competition with decay by emission of a photon into many other field modes. Because the density of states for the electromagnetic field (modes per unit volume and unit energy interval) scales as ω^3 with the

photon energy $\hbar\omega$, this is a very stiff competition at x-ray photon energies. For an estimate, we assume comparable oscillator strenghts for the 1st-photon (decay to ground state) and 2nd-photon (further excitation) processes, and then compare directly the number of modes. The incident (2nd-photon) x-rays are present in one mode given by a quantization volume $\mathcal{V}$ of, say, 10^{-22} m^3 (a focus with a 10-nm Airy disc and a coherence length of a typical x-ray linewidth of 1 eV). The number of modes available for spontaneous emission is given by $\frac{4}{3}\pi k^2 \Delta k \mathcal{V}$, where k is the wave number ($k = 5 \cdot 10^{10}$ m^{-1} at 10 keV), and $\Delta k = 5 \cdot 10^5$ m^{-1} is given by the same 1-eV linewidth ($\Delta k \mathcal{V}$ is actually independent of the linewidth). So, in this example, there are $5 \cdot 10^5$ modes available for spontaneous emission compared to 1 of the incident x-rays. In the context of the Wigner-Weisskopf theory of spontaneous emission, this is the same as saying that a second photon has to be absorbed within the short lifetime of the intermediate excited state, which is typically in the order of femtoseconds for hard x-rays.

For coherent nonlinear scattering of a wave a in the presence of another of intensity I_b in a sample of free electrons, one can calculate [1] a cross section per electron of the order of $I_b r_e^4/(\hbar\omega^2)$.

4 Nonlinear Optics of Free Electrons, Parametric Down Conversion

An x-ray nonlinear-optical experiment that is feasible - and has been done - at a 3rd-generation source is parametric down conversion of x-rays (XPDC). This is the effect of a photon decaying spontaneously into a pair of highly correlated, and possibly entangled, photons in a nonlinear optical medium. It can be understood as four-wave mixing driven by vacuum fluctuations. The converter medium is a crystal of some light element, such as diamond. The binding energies of the constituent electrons is much lower than the x-ray photon energy, and can be considered free electrons undergoing transverse oscillations under the influence of the electric field of one wave. The magnetic field of another wave can then induce longitudinal oscillations through the Lorentz force, which can emit into the directions of other, diffractively coupled waves - hence the need for a crystal. This extremely weak nonlinear response is somewhat offset by the high modal density at x-ray photon energies, i.e, strong vacuum fluctuations driving the process. Even so, XPDC is a weak effect, and the highest event rate observed [1] to date is about 1 in 10 s at an incident x-ray flux of 10^{12} per second.

5 Nuclear γ-Ray Superradiance

Another quantum-optical effect that can be - and has been - observed without an XFEL is nuclear γ-ray superradiance, i.e., collective spontaneous emission from an ensemble of nuclei resonantly excited by a pulse of synchrotron radiation. Its signature phenomena are speedup and directionality of emission [2, 3]. In all nuclear-resonance experiments performed to date, it is a single-photon process, and thus a purely quantum-optical phenomenon. An example of the appliation of quantum-optical concepts to x-ray science is the possibility of controlling superradiant emission in space and time by manipulation of the ensemble coherence. One could, for example, introduce spatial and temporal patterns of accumulated dynamic phases through energy-level shifts due to transient magnetic fields. These will lead to redirection of the superradiant emission at a suitably chosen time.

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